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The influence of nanoscale inorganic content over optical and surface properties of model composites

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ABSTRACT

Objectives: To investigate the influence of nanoscale inorganic content over optical and surface properties of model composites before and after ageing.
Methods: Three model composites were formulated with silica fillers in nanoscale of 7 nm (G1), 12 nm (G2) and 16 nm (G3), at 45.5% by weight in a matrix of BisGMA/TEGDMA 1:1. Color coordinates (CIE $L^*a^*b^*$ parameters), color difference (ΔE^*), translucency parameter (TP), surface gloss (SG) and surface roughness (SR) were measured before and after ageing procedures of immersion in water and toothbrush abrasion. Surface hardness (SH) were evaluated before and after immersion in absolute ethanol. Results were submitted to two-way ANOVA followed by Tukey's post hoc test performed at a pre-set alpha of 0.05.
Results: Regarding CIE $L^*a^*b^*$ parameters, a darkening, a redness and a blueness effect, were respectively detected after water storage for all groups. Smaller filler sizes (G1) had the highest CIE b^* values, whereas medium (G2) ($p < 0.05$) had higher values than larger fillers (G3) ($p < 0.05$) either before or after water immersion. Toothbrush abrasion did not produce any significant effect on CIE a^* and CIE b^* values, however increased CIE L^*, decreased TP in addition to produce rougher and matte surfaces in all groups. Filler size did not influence ΔE^* ($p > 0.05$), although a tendency towards lower values in smaller filler materials was observed. Ageing after immersion in absolute ethanol decreased SH for all model composites.
Conclusions: Filler sizes and ageing procedures influenced the optical and surface properties of the nanostructured composites evaluated in this study.
Clinical significance: Filler size influenced optical and surface properties of resin composites. Materials containing smaller filler size offered improved optical stability and surface properties that can lead to longer maintenance of the restoration's appearance in the oral environment.

1. Introduction

Color and appearance of teeth and restorative materials are a complex phenomenon influenced by several factors such as lighting conditions, translucency, opacity, scattering and light transmission, brightness, fluorescence and opalescence. The surface texture and the size of a restoration may also produce an effect on the perception of light by the observer.[1–4] It is known that the surface of the restoration can be polished to a high gloss.[5,6] Being an attribute of visual appearance, gloss is directly influenced by the surface roughness[7] as originates from a geometrical distribution of light reflected by the surface.[8] Changes in surface gloss in the oral environment due

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to mechanical wear and/or chemical degradation of composite restorations may result in unfavourable aesthetics over time.

When incident light strikes a materials' surface, part of the light is reflected at the boundary between the material and air. The other part of the light is absorbed and interactions such as: reflection, refraction, absorption, scattering and/or transmission occur throughout the restorative composite material. This happens because of changes in the refractive index, once the ratio of the speed of light in a vacuum is different from the speed of light inside.[9] The back of the mouth receives little light and appears dark. As a consequence, it is not uncommon to observe in Class III or IV restorations a grey effect in the composite restoration compared with the color of the remaining tooth tissue. This occurs if translucent materials are placed in a cavity with no opaque structure backing, affected by the dark background the restoration will appears dark or grey.[10] Thus, translucency and opacity are fundamental characteristics in order to achieve aesthetic restorations with composites, since they are indicators of the quality and quantity of reflected light.[11] The main component of a dental composite that significantly affects the translucency is its inorganic content.[12] Moreover, the quantity, the size and the shape of the fillers affect the light scattering in dental composites.[13]

In the last decade several dental resin–composites were introduced utilising nano-filler technology and thus classified as nanocomposites or nanohybrids. The nano-fillers can be dispersed in high concentrations within the organic matrix, improving physical and mechanical properties of the composites after setting.[11,14–17] Defined as ''*nanocomposites*'' these materials appear to bring benefits such as improved surface smoothness,[15,18] excellent resistance to mechanical wear, high aesthetics due to the higher volume fraction of loads and ease of maintenance and polishing.[19,20] Thus, strongly indicated for restorations in areas where aesthetics are indispensable.

Surface texture and optical properties can be modified by several factors such as wear of fillers, degradation of the organic matrix or weakening of organic matrix-filler bonding.[21] Some studies have investigated the influence of the type, size and volume of fillers in the optical properties of experimental dental composites.[17,22–24] Nevertheless, there is no data in the literature about the influence of size and volume of fillers in nanostructured composites on optical and surface properties of the materials when they are submitted to ageing procedures. In this study, a series of model resin-composites with systematically varied filler sizes was examined. The aim was to investigate the effect of filler-size on (i) surface properties such as roughness (SR), surface gloss (SG) and hardness (SH) and (ii) optical properties such as color measurements (CIE $L^*a^*b^*$ parameters), color difference (ΔE^*) and translucency parameter (TP) before and after ageing procedures. The hypotheses tested were that filler-size and ageing procedure would affect: (i) optical properties (ii) surface properties.

2. Materials and methods

2.1. Materials formulation

The monomers 2,2 bis[4-2(2-hydroxy-3-methacroyloxypropoxy)phenyl] propane (Bis-GMA, Esstech, Essington, PA,USA) and triethyleneglycol dimethacrylate (TEGDMA, Esstech, Essington PA, USA) were mixed in equal parts by weight. Camphorquinone (0.5% mol; ESSTECH, Essington, PA, USA) and ethyl 4-dimethylaminobenzoate (1% mol; Sigma–Aldrich, Chemie, Steinheim, Germany) were added as photoinitiators. The photoinitiator/co-initiator ratio used was based on a previous paper.[25] After this, three groups were created by the addition of the following silane-treated fumed silica nanofillers:

G1) 7 nm average diameter (R812 - Evonik, Degussa, Germany);
G2) 12 nm average diameter (R805 - Evonik, Degussa, Germany) and
G3) 16 nm average diameter (R972 - Evonik, Degussa, Germany).

The smaller the filler size the more difficult it becomes its incorporation into matrix. Fillers were added at 45.5 total wt% into the organic matrix. This was the maximum wt% achieved by hand-mixed incorporation for the group formulated with 7 nm diameter fillers (G1). Thus, in order to keep standardisation, the same filler amount (45.5%) was incorporated in the others two model materials (G2 and G3). Table 1 describes formulation of model composites used.

Six disc specimens (8 mm × 2 mm) were prepared from each material. The samples were irradiated for 30 s with a light curing unit (Radii CAL–SDI, USA) operated in standard mode and emitting 1000 mW/cm^2 irradiance, as measured with a hand-held radiometer. Specimens were then submitted to mechanical polishing in a grinding/polishing machine (Buehler, Lake Bluff, USA) with a sequence of 2000-grit and 4000-grit SiC papers under continuous water cooling.

2.2. Ageing procedures

Optical and surface properties were evaluated initially 24 h after curing and dry storage. The specimens were then stored in distilled water at 37 °C and measurements were repeated after 10, 20, 30 and 60 days of water immersion. After 60 days of water immersion, samples were submitted to toothbrush abrasion.

A toothbrush simulating machine (MEV2 – Odeme, Brazil) was used to abrade the samples' surfaces. The toothbrush machine had six separate ''stations'' and separate toothbrush holders which were driven by a motor. Hence six specimens were concurrently but individually subjected to an equal amount of toothbrush/dentifrice abrasion during each testing period. Soft toothbrushes (Dr. Veit Soft, Dr. Veit Produtos Odontológicos, Brazil) were fixed in a toothbrush holder so that all the bristles were in contact with the specimen. The testing machine was adjusted to apply 2.5 N vertical load on the specimen during horizontal movement of the toothbrush throughout the test. A current dentifrice (Colgate Total, Colgate-Palmolive, Manchester, UK) was used to form the slurry (2:1, water:toothpaste) according to ISO/TS 1469-1. Each ''station'' of the machine was filled with 5 ml of slurry. All specimens were brushed with inverse strokes 20,000 times, as measured with an incorporated metre. This corresponds to approximately 2 years of toothbrushing.[26] The toothbrushes and the slurry were replaced for each specimen every 2500 cycles. After abrasion, specimens were removed from the

Table 1 – Formulation of model composites evaluated.

	Average filler size	Type of filler	Filler fraction (wt%)	Matrix fraction (wt%)	Matrix formulation
G1	7 nm (Evonik R812)	Fumed Silica (SiO_2)	45.5	54.5	BisGMA (64.5%) + TEGDMA (35.5%) CQ (0.5 mol%) + EDMAB (1.0 mol%)
G2	12 nm (Evonik R805)	Fumed Silica (SiO_2)	45.5	54.5	BisGMA (64.5%) + TEGDMA (35.5%) CQ (0.5 mol%) + EDMAB (1.0 mol%)
G3	16 nm (Evonik (R972)	Fumed Silica (SiO_2)	45.5	54.5	BisGMA (64.5%) + TEGDMA (35.5%) CQ (0.5 mol%) + EDMAB (1.0 mol%)

machine, rinsed with tap water, cleaned in an ultrasonic bath with distilled water for 5 min, and gently air dried. The measurements of optical and surface properties were repeated.

For surface hardness (SH) evaluation, all specimens were measured initially 24 h after curing and dry storage and after immersion in absolute ethanol for 7 days.

2.3. Optical properties measurements

Color was measured according to the CIE $L^*a^*b^*$ parameters in the reflectance mode, with the SCI mode,[27] over a zero calibrating box (CIE $L^* = 0.0$, $a^* = 0.0$, and $b^* = 0.0$), a white background (CIE $L^* = 93.2$, $a^* = -0.3$, and $b^* = 1.6$) and a black background (CIE $L^* = 0.5$, $a^* = 0.7$, and $b^* = -0.6$) using a spectrophotometer with illuminating/measuring geometry d/8° (CM-2600d, Konica Minolta, Japan). The size of measuring aperture used was 3 mm of diameter. Illuminating and viewing configurations complied with CIE 10° observer geometry and D65 illuminant. An average of three readings (in the centre of top surface) for each sample was calculated. CIE L^*, a^*, and b^*-values were automatically calculated by the machine. The CIE L^* axis is the lightness, where 100 is white and 0 is black. The axes CIE a^* and CIE b^* are the red-green and yellow-blue chromatic coordinates. A positive CIE a^* or CIE b^* value represents a red or a yellow shade, respectively. Measurements were obtained 24 h after polymerisation under dry and dark conditions and after immersion in distilled water for 10, 20, 30 and 60 days, and after toothbrush abrasion (20,000 brushing cycles).

The color changes, ΔE^*, were calculated from the single color values CIE L^*, CIE a^* and CIE b^* according to the following formula:

$$\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2},$$

where ΔL^*, Δa^* and Δb^* represents the mathematical differences between each parameter (CIE L^* CIE a^* or CIE b^*) at different periods of time compared with initial values (24 h after curing).

Translucency Parameter (TP) was calculated by using the formula:

$$TP = [(L^*_w - L^*_b)^2 + (a^*_w - a^*_b)^2 + (b^*_w - b^*_b)^2]^{1/2},$$

where the subscript "w" refers to CIE $L^*a^*b^*$ parameters for each specimen on the white background and the subscript "b" refers to values for specimen on the black background.

2.4. Surface properties analysis

2.4.1. Surface roughness

A surface profiler (SJ-201 – Mitutoyo, Japan) was used to measure the surface roughness. The diamond stylus was traversed at a constant speed of 1 mm/s across the surface with a force of 6 mN. Six line scans were performed per specimen surface, three in horizontal and three in perpendicular directions. The cut-off length was 0.25 mm and the measuring length 2 mm. An average of six readings per specimen was used. The amplitude parameter R_a, which relates the arithmetic mean between the peaks and valleys of the surface, was used.

2.4.2. Surface gloss

The gloss was measured with a glossmeter (ZGM 1110 – Zehntner, Switzerland) which was calibrated against a black glass standard provided by the manufacturer. Measurements were performed at 60° light incidence and reflection angles relative to the vertical axis.[8,19,28–30] The measuring window was 4.7 mm × 2 mm.

2.4.3. Surface hardness

Knoop hardness values was measured on the irradiated surfaces using an indenter (Micromet 5104 - Buehler, Japan) under a load of 25 g applied for 15 s. Measurements were performed at five locations on each specimen, and the average Knoop Hardness Number (KHN) was recorded as the hardness for a given specimen. The measurements were taken initially after curing and repeated after 7 days immersion in absolute ethanol.[31,32]

Data were submitted to two-way analysis of variance (ANOVA) followed by Tukey's test, at a 5% global level of significance.[33]

3. Results

Fig. 1 presents values of CIE L^*, CIE a^* and CIE b^* of each group, over time on water storage and after the toothbrush abrasion. For all groups immersion in water for 60 days significantly decreased CIE L^* values ($p < 0.05$). On the other hand, after toothbrush abrasion CIE L^* values increased ($p < 0.05$) compared with immersion for 60 days in water. Materials formulated with larger filler size (G3) revealed higher CIE L^* values ($p < 0.05$) than medium and smaller filler sizes (G2 and G1). Although, there was an increase in CIE a^* values after immersion in water for 60 days in all groups evaluated, there was no statistical difference among them ($p > 0.05$). On the other hand, there was a significant decrease in CIE b^* values after immersion in water for 60 days for all groups evaluated ($p < 0.05$) affected by the filler sizes. According to the results G1 had the highest CIE b^* values, whereas G2 ($p < 0.05$) had higher values than G3 ($p < 0.05$) either before or after water immersion. Thus, the smaller the filler size the greater the yellowing effect, which in turn was reduced with water

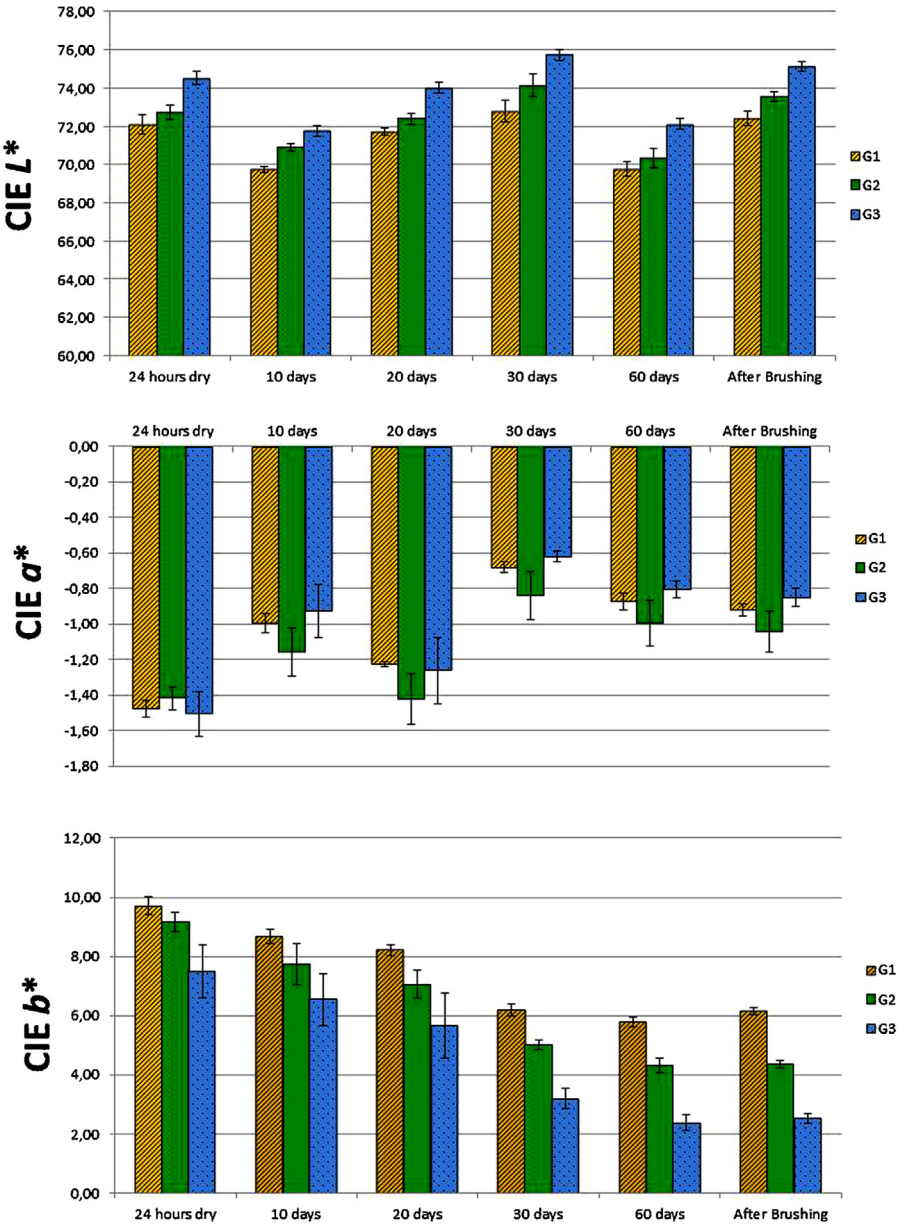

Fig. 1 – CIE $L^*a^*b^*$ values of model composites over time of water storage and abrasion.

storage. Toothbrush abrasion did not produce any statistically significant effect on CIE a^* and CIE b^* values.

Fig. 2 displays ΔE^* and TP values after ageing procedures for each material. Filler size did not influence the color difference ($p > 0.05$), although a tendency was observed to lower ΔE^* values in smaller filler materials (G1). Water immersion and toothbrush abrasion affected ΔE^* values ($p < 0.05$). In all groups TP values decreased after ageing procedures ($p < 0.05$) and it was not affected by the filler size ($p > 0.05$).

All materials exhibited very smooth surfaces before toothbrush abrasion. R_a values ranged from 0.03 to 0.06 μm and 0.76 to 0.90 μm before and after ageing respectively. Surface roughness (R_a parameter) was influenced by the filler size. According to the initial values (24 h after curing) larger filler size (G3) presented rougher surfaces than medium (G2) and smaller (G1) filler materials ($p < 0.05$). After water storage R_a values increased for all materials with no significant statistical difference among the groups. Nevertheless, after toothbrush abrasion an exponential increase of surface roughness values were observed among filler size. The larger filler sizes also revealed higher surface roughness values followed by the medium and smaller fillers ($p < 0.05$).

Surface properties results are presented in Fig. 3. Gloss values ranged between 67.5 and 92.0 GU before abrasion and between 40.0 and 48.8 GU after toothbrush abrasion. For all materials a statistically significant reduction in gloss was observed after immersion in water ($p < 0.05$) and after toothbrush abrasion ($p < 0.05$). Gloss retention comparison among the different materials was performed by determining the percentage change in gloss for a given material between

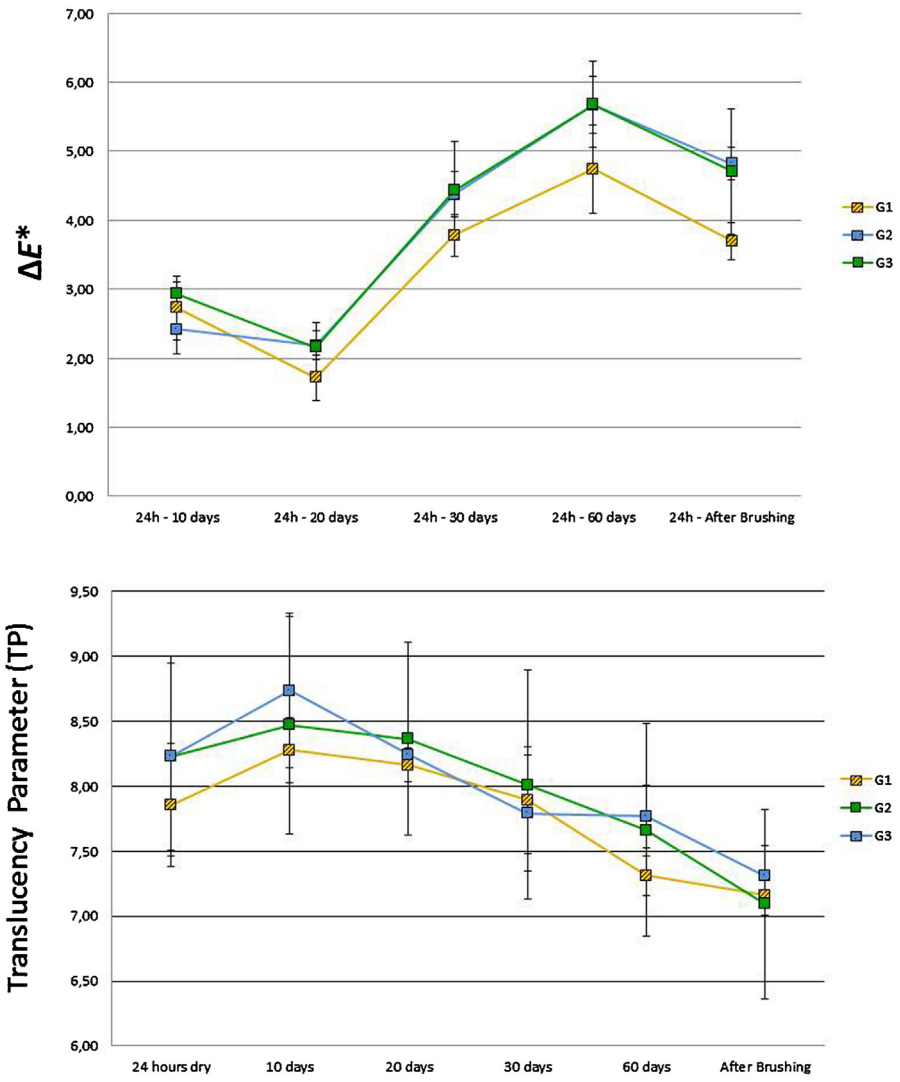

Fig. 2 – ΔE* and translucency parameter (TP) values of model composites over time of water storage and abrasion.

the immediate reading and after ageing procedures. According to this percentage, composites formulated with larger filler sizes (47.8%) presented less gloss retention than medium (46.9%) and smaller filler (40.7%) composites.

Regarding surface hardness, after 7 days immersion in absolute ethanol, all materials were significantly softer than after 24 h after curing. Before and after immersion larger filler size presented higher KHN values ($p < 0.05$) than medium and smaller filler size which produced similar outcomes.

4. Discussion

Nano filler size composites may have optical features that promote aesthetic matching of restorative materials with surrounding dental hard tissues. In comparison with hybrid resin-composites, nanofillers are believed to offer improved mechanical and optical properties.[34] However, there is no consensus on the benefits of these materials, especially in relation with their long term clinical performance.

Optical properties were influenced by ageing procedures and filler size, thus first hypothesis was accepted. Immersion in water for 60 days produced significant changes in CIE $L^*a^*b^*$ parameters. For the CIE L^* values were reduced after water storage in all materials evaluated. This darkening effect produced by water remained stable from day 10 up to day 60 of evaluation, which might be related to the fact that water sorption could have stabilized after 10 days. For the evaluation of CIE a^* and CIE b^* parameters, a redness and a blueness effect, were respectively detected after water storage in all groups. In contrast, toothbrush abrasion produced an increase in lightness (CIE L^*) compared with initial measurements and did not produce any changes in CIE a^* and CIE b^* values. In G1 and G2 the darkening and yellowing effects were more evident than in G3. Changes in CIE $L^*a^*b^*$ parameters due to water storage could be explained by hydrolysis.

Color stability is critical to the long-term clinical aesthetic performance of restorations. Regarding ΔE* values, after water immersion values increased in all groups but were not affected

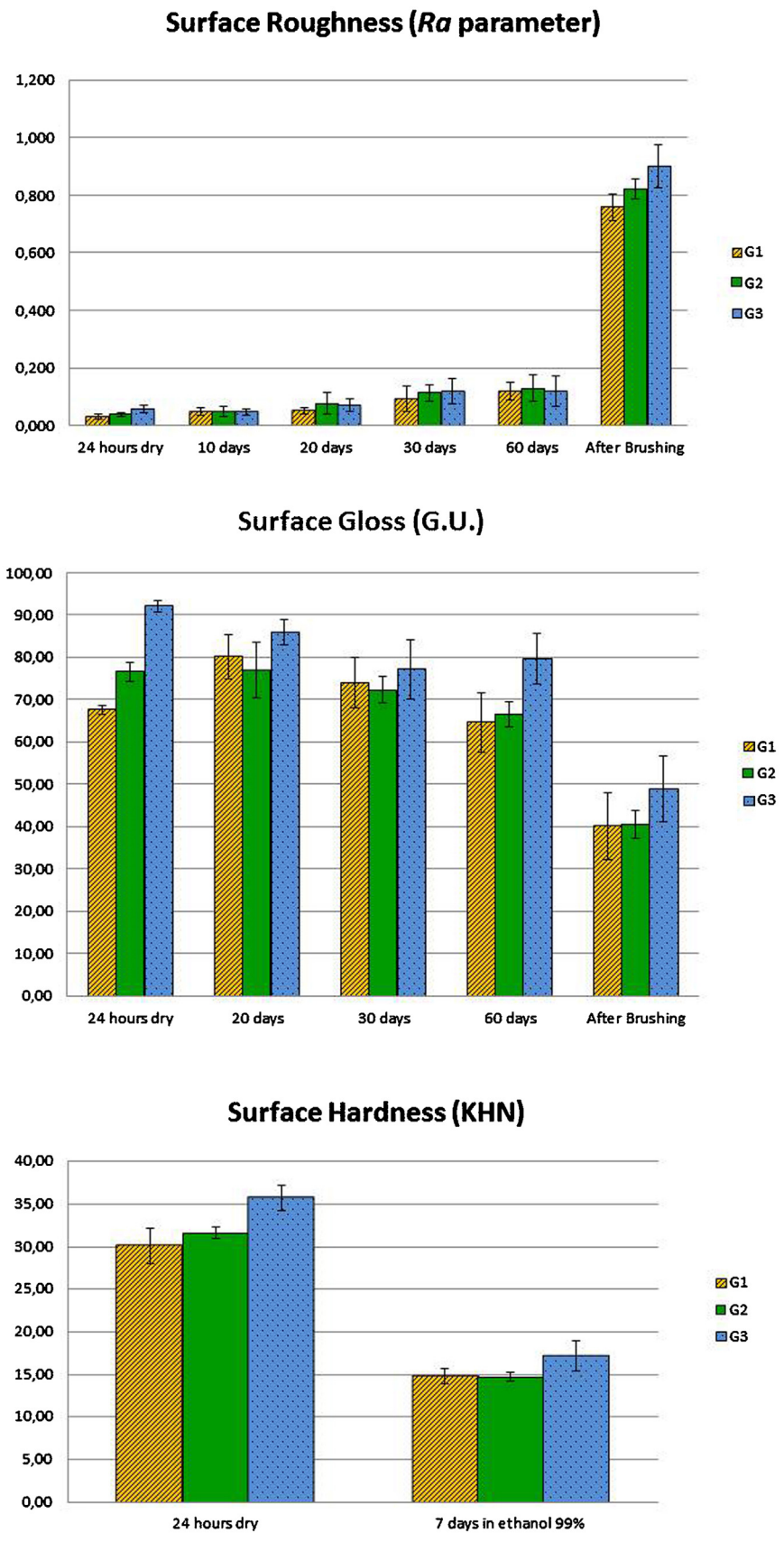

Fig. 3 – (A) Surface roughness, (B) surface gloss and (C) surface hardness values of model composites over ageing procedures.

by filler size. G1 showed a tendency of better color stability and presented lower ΔE^* values ($\Delta E^* = 4.7$ after 60 days in water and $\Delta E^* = 3.6$ after abrasion). This can be explained by a more homogeneous distribution of the fillers of the matrix resin, compared with larger fillers, and also by the process of silanisation.[35] Color changes in composite materials may be caused by intrinsic and extrinsic factors. Intrinsic factors such as the alteration of the organic matrix and/or at the interface between matrix and fillers by water seems to influence ΔE^* values over time. It has been shown in the literature that ΔE^* values above 1 are perceptible with the naked eye[36] and ΔE^* values similar or superior to 3.3 are considered clinically unacceptable.[37,38] The ΔE^* values, in this study, ranged between 3.0 and 5.7. The color changes observed are due to colored oxidation products forming from the amine co-initiator. The more the filler scatters light, the less noticeable and measurable is this color change, primarily in CIE b^*. The smaller the scattering centres, the less the light is scattered in the visible range. Still, all of the filler sizes used were in a range (7, 12, and 16 nm) well below the visible range (>400 nm) and thus ''nominal'' size wouldn't be expected to have an effect on visible-range scattering.

If there is a mismatch in refractive indices between filler and matrix, the filler will increase light scattering resulting in opaque materials. If the opacity of composite increases, the refractive index difference between filler and organic matrix also increases.[34] Thus, among several factors, filler sizes may affect translucency of dental composite materials. If filler size is far below the wavelength of visible light, it will not scatter the light or absorb the light, resulting in the human eye's inability to detect the fillers. Thus, dental composites with nano-fillers produce superior translucency and deliver optimal aesthetics.[39] After water immersion and toothbrush abrasion the translucency parameter was significantly decreased, however no differences among filler sizes were observed. As reported in others researches,[10,40] the CIEL$^*a^*b^*$ values tended to reduce when the materials were placed over a black background in comparison with a white background. The black background may be used to simulate the dark background of the mouth in extensive Class III or IV restorations. For this type of cavities little or no existing tooth structure surrounds the restoration to provide a base for reflected or transmitted color. Composites show more dark, greenish and bluish aspects over a black background, which explains the grey effect in some extensive Class III or IV restorations.[10]

Filler sizes and ageing procedures produced significant changes on surface properties; hence the second hypothesis was accepted. Water immersion and toothbrush abrasion increased surface roughness values in all groups. G3 had the rougher surfaces and showed a larger increase in R_a values than G2 and G1 after ageing procedures. In resin-composites, abrasion commonly takes place through a gradual removal of the organic matrix. This eventually leaves the fillers unsupported and susceptible to exfoliation.[41] As a result, when larger filler particles are plucked out, a rougher surface is expected. Therefore, materials with smaller filler sizes were more resistant to toothbrush abrasion. This could be related to their inter-filler spacing, which increases as filler size increases, thus decreasing the inter-filler spacing was the key for improving wear-resistance of composites.[42] Fillers in close proximity protect the softer resin-matrix from abrasives and produce a larger contact area between fillers and the antagonist, thus reducing wear.[43] Toothbrush abrasion of composite materials varies in accordance with the type of composite,[29] type of toothpaste,[44] and the nature of toothbrush employed.[45] The toothbrush was kept in contact with the specimens with a standardized force through a toothbrush holder. As matrix was the same for all groups, the only variable influencing the results was the inorganic content of model composites.

Differences in gloss between a restoration and surrounding enamel are clinically relevant as the human eye can easily detect differences in gloss even if their color is matched.[45] On the other hand, high gloss reduces the effect of a color difference, since the color of reflected light is predominant rather than the color of underlying composite material.[46] The angle of illumination affects the amount of reflected light. According to Silikas et al.[47] 60° light incidence is considered more reliable from a clinical perspective, since it is closer to the perception of tooth gloss by an observer. Higher gloss values indicate smooth and high-lustre surfaces.[48] For all materials tested the surface became statistically less glossy after ageing procedures, but without a correlation with the size of fillers ($r = 0.340$; $p = 0.371$). Light reflectivity seems, therefore, to be related to mean filler size and to homogeneity of the filler–matrix complex. According to Lee et al.[29] larger fillers and lower homogeneity of the filler-matrix complex results in inferior light reflectivity. Although, G1 and G2 presented lower values of gloss units than G3, these groups presented higher percentage of gloss retention. In other words, this outcome means that the larger the filler size, the lower the gloss retention after ageing procedures. The decrease in gloss, was reported in others studies with commercial composites[26,45,49] and there was a negative correlation with surface roughness.

Regarding the surface hardness, immersion in absolute ethanol resulted in a significant decrease of surface hardness for all groups. Due to the higher amount of [–OH] present in the ethanol, a higher absorption occurs by the polar portion of the matrix, causing swelling of the material. This dimensional change in the matrix causes stress at the matrix–silane–filler particle interfaces, resulting in degradation of the bond. As a consequence, inorganic particles detach from the surface, causing an increase in surface roughness and consequently reduction on the surface hardness of the specimens.[50] Either before or after ageing procedures, larger filler sizes presented higher KHN values compared to medium and smaller filler materials. As proposed by Moraes et al.[51] it seems that the larger the filler particle, the higher the resistance of surface to indention.

In general, nano-filler structured composites are very difficult to process with conventional techniques. There is a strong tendency of fillers to agglomerate, which is hard to overcome by limiting the shear force during compounding. The smaller the filler, the more difficult it becomes to break down these agglomerates to result in a homogeneous distribution within the organic matrix.[52] In this study the inorganic filler and the organic matrix were manually hand-mixed and there was a possibility to generate clusters inside

the inorganic phase as well as not-equally distributed filler within the material, affecting how light interacts with the composite. Although all care was taken to standardize all factors involved in the current study, the used fillers have different silane coverages and were used as received. This is important to be considered since the silane layer may also influence the final optical properties.[53] Additional studies have now been conducted by this research group in order to elucidate the interaction between the filler size and the silane treatments over the final optical properties.

Previous studies have used commercially available composites to study the influence of the filler size on the optical properties. In example, Janus et al.[54] studied the surface roughness and morphology of a hybrid composite (Tetric Ceram, Ivoclar) and three nanocomposites (one ''pure'' nanofilled composite, Filtek Supreme/3MESPE; and two ''nano-hybrid'' composites, Grandio/Voco; Synergy D6/Coltène) and verified a positive correlation between the average filler size and the surface roughness. On the opposite way, Berger et al.[55] investigated the influence of filler size and finishing systems on the surface roughness and staining of three commercial dental composites (''nanofilled'' Filtek Supreme/3MESPE; ''minifill'' Esthet-X/Dentsply, and ''microfill'' Renamel Microfill/Cosmedent) and concluded that the smallest filler size does not necessarily result in a low surface roughness and staining susceptibility. In the same direction, Gönülol and Yilmaz[56] evaluated the effects of different finishing and polishing techniques on the surface roughness and color stability of commercial composites (two ''nanohybrid'' composites, Grandio/Voco and Aelite Aesthetic/Bisco, two ''nanofillied'' composites, Filtek Supreme XT/3MESPE – Dentine and Translucent, and a ''microhybrid'' composite, Filtek Z250/3M/ESPE) and concluded that composites with smaller filler size did not necessarily show lower surface roughness and discoloration than the others and, furthermore, the authors stated that staining of composite resins was dependent on monomer structure and surface irregularities. As it can be seen, studies that use commercially available composites have found conflicting results and this may be expected since such materials do not differ only in the filler content. Differently, the model composites evaluated in the current research were a solely a mixture of organic monomers, inorganic fillers surrounded by a coupling agent and a photo-initiator/co-initiator system. Therefore, the major contribution is the fact that the main factor influencing the results – regarding the materails' composition – was filler size.

5. Conclusions

Within the limitations of the current study, the authors conclude that:

- Filler sizes influenced the optical and surface properties of the nanostructured composites evaluated in this study.
- Distinct advantages of smaller and medium up to larger fillers were shown regarding to color difference, gloss retention and surface roughness;
- All materials evaluated were affected by ageing procedures. Optical and surface properties were adversely affected by immersion in water and toothbrush abrasion.

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REFERENCES

1. Arikawa H, Fujii K, Kaine T, Inoue K. Light transmittance characteristics of light-cured composite resins. *Dental Materials* 1998;**14**:405–11.
2. Joiner A. Tooth colour: a review of the literature. *Journal of Dentistry* 2004;**32**:3–12.
3. Nakajima M, Arimoto A, Prasansuttiporn T, Thanatvarakorn O, Foxton RM, Tagami J. Light transmission characteristics of dentine and resin composites with different thickness. *Journal of Dentistry* 2012;**40**:e77–e82.
4. Li R, Ma X, Liang S, Sa Y, Jiang T, Wang Y. Optical properties of enamel and translucent composites by diffuse reflectance measurements. *Journal of Dentistry* 2012;**40**:e40–e47.
5. Lu H, Roeder LB, Lei L, Powers JM. Effect of surface roughness on stain resistance of dental resin composites. *Journal of Esthetic and Restorative Dentistry* 2005;**17**:102–8.
6. Antonson SA, Yazici AR, Kilinc E, Antonson DE, Hardigan PC. Comparison of different finishing/polishing systems on surface roughness and gloss of resin composites. *Journal of Dentistry* 2011;**39**:e9–17.
7. Van Dijken JW, Ruyter IE. Surface chracteristics of posterior composites after polishing and toothbrushing. *Acta Odontologica Scandinavica* 1987;**45**:337–46.
8. Heintze SD, Forjanic M, Rousson V. Surface roughness and gloss of dental materials as a function of force and polishing time in vitro. *Dental Materials* 2006;**22**:146–65.
9. Paravina RD, Powers JM. Esthetic color training in dentistry. Philadelphia: Elsevier Mosby; 2004.
10. Ikeda T, Murata Y, Sano H. Translucency of opaque–shade resin composites. *American Journal of Dentistry* 2004;**17**:127–30.
11. Kim KH, Ong JL, Okuno O. The effect of filler loading and morphology on the mechanical properties of contemporary composites. *Journal of Prosthetic Dentistry* 2002;**87**:642–9.
12. Moharamzadeh K, Azzopardi N, Wood DJ, Martin N, van Noort R. Effect of resin matrix composition on the translucency of experimental dental composite resins. *Dental Materials* 2009;**25**:1564–8.
13. Lee YK. Influence of filler on the difference between the transmitted and reflected colors of experimental resin composites. *Dental Materials* 2008;**24**:1243–7.
14. Terry DA. Direct applications of a nanocomposite resin system. Part 1. The evolution of contemporary composite materials. *Practical Procedures & Aesthetic Dentistry* 2004;**16**:417–22.
15. Chen MH. Update on dental nanocomposites. *Journal of Dental Research* 2010;**89**:549–60.
16. Hosoya Y, Shiraishi T, Oshiro M, Ando S, Miyazaki M, García-Godoy F. Color characteristics of resin composites in different color modes and geometries. *Journal of Oral Science* 2009;**51**:123–30.

17. Lim YK, Lee YK, Lim BS, Rhee SH, Yang HC. Influence of filler distribution on the color parameters of experimental resin composites. *Dental Materials* 2008;**24**:67–73.
18. Turssi CP, Ferracane JL, Serra MC. Abrasive wear of resin composites as related to finishing and polishing procedures. *Dental Materials* 2005;**21**:641–8.
19. Cavalcante LM, Masouras K, Watts DC, Pimenta LA, Silikas N. Effect of nanofillers' size on surface properties after toothbrush abrasion. *American Journal of Dentistry* 2009;**22**:60–4.
20. Mitra SB, Wu D, Holmes BN. An application of nanotechnology in advanced dental materials. *Journal of the American Dental Association* 2003;**134**:1382–9.
21. Kaizer MR, Diesel PG, Mallmann A, Jacques LB. Ageing of silorane-based and methacrylate-based composite resins: effects on translucency. *Journal of Dentistry* 2012;**40**:e64–71.
22. Dos Santos GB, Alto RV, Filho HR, da Silva EM, Fellows CE. Light transmission on dental resin composite. *Dental Materials* 2008;**24**:571–6.
23. Emani N, Sjodahl M, Soderholm KJ. How filler properties, filler fraction, sample thickness and light source affect light attenuation in particulate filled resin composites. *Dental Materials* 2005;**21**:721–30.
24. Miyagawa Y, Powers JM, O'Brien WJ. Optical properties of direct restorative materials. *Journal of Dental Research* 1981;**60**:890–4.
25. Schneider LF, Cavalcante LM, Consani S, Ferracane JL. Effect of co-initiator ratio on the polymer properties of experimental resin composites formulated with camphorquinone and phenyl-propanedione. *Dental Materials* 2009;**25**:369–75.
26. Tanoue N, Matsumura H, Atsuta M. Wear and surface roughness of current prosthetic composites after toothbrush/dentifrice abrasion. *Journal of Prosthetic Dentistry* 2000;**84**:93–7.
27. Hosoya Y, Shiraishi T, Oshiro M, Ando S, Miyazaki M, Powers JM. Effects of specular component on color differences of different filler type resin composites after aging. *Journal of Dentistry* 2009;**37**:585–90.
28. Paravina RD, Roeder L, Lu H, Vogel K, Powers JM. Effect of finishing and polishing procedures on surface roughness, gloss and color of resin-based composites. *American Journal of Dentistry* 2004;**17**:262–6.
29. Lee YK, Lu H, Oguri M, Powers JM. Changes in gloss after simulated generalized wear of composite resins. *Journal of Prosthetic Dentistry* 2005;**94**:370–6.
30. Waheeb N, Silikas N, Watts D. Initial polishing time affects gloss retention in resin composites. *American Journal of Dentistry* 2012;**25**:303–6.
31. Schneider LF, Moraes RR, Cavalcante LM, Sinhoreti MA, Correr-Sobrinho L, Consani S. Cross-link density evaluation through softening tests: effect of ethanol concentration. *Dental Materials* 2008;**24**:199–203.
32. Voltarelli FR, dos Santos-Daroz CB, Alves MC, Cavalcanti NA, Marchi GM. Effect of chemical degradation followed by toothbrushing on the surface roughness of restorative composites. *Journal of Applied Oral Science* 2010;**18**:585–90.
33. Hannigan A, Lynch CD. Statistical methodology in oral and dental research: Pitfalls and recommendations. *Journal of Dentistry* 2013;**41**:385–92.
34. Kim JJ, Moon HJ, Lim BS, Lee YK, Rhee SH, Yang HC. The effect of nanofiller on the opacity of experimental composites. *Journal of Biomedical Materials Research Part B Applied Biomaterials* 2007;**80 B**:332–8.
35. Sideridou ID, Karabela MM. Sorption of water: ethanol or ethanol/water solutions by light-cured dental dimethacrylate resins. *Dental Materials* 2011;**27**:1003–10.
36. Kuehni FG, Marcus RT. An experiment in visual scaling of small color differences. *Color Research Application* 1979;**4**:83–91.
37. Ruyter IE, Nilner K, Moller B. Color stability of dental composite resin materials for crown and bridge veneers. *Dental Materials* 1987;**3**:246–51.
38. Johnston WM, Kao EC. Assessment of appearance match by visual observation and clinical colorimetry. *Journal of Dental Research* 1989;**68**:819–22.
39. Ure D, Harris J. Nanotechnology in dentistry: reduction to practice. *Dental Update* 2003;**30**:10–5.
40. Ikeda T, Sidhu SK, Omata Y, Fujita M, Sano H. Colour and translucency of opaque-shades and body-shades of resin composites. *European Journal of Oral Sciences* 2005;**113**:170–3.
41. Condon JR, Ferracane JL. In vitro wear of composite with varied cure, filler level, and filler treatment. *Journal of Dental Research* 1997;**76**:1405–11.
42. Turssi CP, Ferracane JL, Vogel K. Filler features and their effects on wear and degree of conversion of particulate dental resin composites. *Biomaterials* 2005;**26**:4932–7.
43. Lim BS, Ferracane JL, Condon JR, Adey JD. Effect of filler fraction and filler surface treatment on wear of microfilled composites. *Dental Materials* 2002;**18**:1–11.
44. Goldstein GR, Lerner T. The effect of toothbrushing on a hybrid composite resin. *Journal of Prosthetic Dentistry* 1991;**66**:498–500.
45. Ardu S, Braut V, Uhac I, Benbachir N, Feilzer AJ, Krejci I. Influence of mechanical and chemical degradation on surface gloss of resin composite materials. *American Journal of Dentistry* 2009;**22**:264–8.
46. O'Brien WJ, Johnston WM, Fanian F. The surface roughness and gloss of composites. *Journal of Dental Research* 1984;**63**:685–8.
47. Silikas N, Kavvadia K, Eliades G, Watts DC. Surface characterization of modern resin composites: a multitechnique approach. *American Journal of Dentistry* 2005;**18**:95–100.
48. Kakaboura A, Fragouli M, Rahiotis C, Silikas N. Evaluation of surface characteristics of dental composites using profilometry, scanning electron, atomic force microscopy and gloss-meter. *Journal of Materials Science Materials in Medicine* 2007;**18**:155–63.
49. Neme AM, Wagner WC, Pink FE, Frazier KB. The effect of prophylactic polishing pastes and toothbrushing on the surface roughness of resin composite materials in vitro. *Operative Dentistry* 2003;**28**:808–15.
50. Schneider L, Moraes R, Cavalcante L, Sinhoreti M, Correr-Sobrinho L, Consan S. Cross-link density evaluation through softening tests: effect of ethanol concentration. *Dental Materials* 2008;**24**:199–203.
51. Moraes RR, Gonçalves LS, Lancellotti AC, Consani S, Correr-Sobrinho L, Sinhoreti MA. Nanohybrid resin composites: nanofiller loaded materials or traditional microhybrid resins? *Operative Dentistry* 2009;**34**:551–7.
52. Rong MZ, Zhang MQ, Pan SL, Friedrich K. Interfacial effects in polypropylene–silica nanocomposites. *Journal of Applied Polymer Science* 2004;**92**:1771–81.
53. Schneider LF, Ferraz L, Rego G, Moraes RR, Cavalcante LM. Dental composite properties' designed by coupling agent content. *Journal of Dental Research* 2012:736. Abstract.
54. Janus J, Fauxpoint G, Arntz Y, Pelletier H, Etienne O. Surface roughness and morphology of three nanocomposites after two different polishing treatments by a multitechnique approach. *Dental Materials* 2010;**26**:416–25.
55. Berger SB, Palialol AR, Cavalli V, Giannini M. Surface roughness and staining susceptibility of composite resins after finishing and polishing. *Journal of Esthetic and Restorative Dentistry* 2011;**23**:34–43.
56. Gönülol N, Yilmaz F. The effects of finishing and polishing techniques on surface roughness and color stability of nanocomposites. *Journal of Dentistry* 2012;**40**:e64–70.